PRNC-202 PUERTO RICO NUCLEAR CENTER RADIONUCLIDE CONTENT OF SOILS FROM BARRIO ISLOTE, 'ARECIBO, PUERTO RICO (JUNE 1976) By Arthur MeB. Block and Richard G. Clements OPERATED BY UNIVERSITY OF PUERTO RICO UNDER CONTRACT NO. B {40-1}1633 FOR US ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION ---Page Break--- Radionuclide Content of Soils from Barrio Islote, Arecibo, Puerto Rico By Arthur MeB. Block and Richard G. Clements Terrestrial Zoology Division Puerto Rico Nuclear Center Caparra Heights Station San Juan, Puerto Rico 00935 ---Page Break--- Chapter I~ Contributors: Chapter II = Contributors: Caveat Lector: Appendix A: Appendix B: ACKNOWLEDGEMENTS A. Introduction B. Qualitative Survey C. Effects on Ecology Terrestrial Zoology Division Arthur MeB. Block Richard G. Clements Félix Santos Ivan Ross Islote Marine Ecology James Parrish Knud Pederson Site Selection Survey Nelson Acosta Elwyn D. Kood C. 37 Estimates Terrestrial Ecology Division Arthur MeB. Block Richard G. Clements On the use of Appendix A for inferences on the geological and petrological development in the northern coastal zone of Puerto Rico. Raw Numerical Spectral Data Chemical Designation of Elements ---Page Break--- Acknowledgments We gratefully acknowledge the Puerto Rico Water Resources Authority (PRWRA), which provided all financial support for personnel, material, and supplies used in bringing this project to fruition. We are particularly indebted to Dr. Juan Bonnet, Mr. Rail McClin, Mr. Rodulfo Gauthier, and Engineers Mejias and Granatges. We wish to thank Dr. L.C. Bate of Oak Ridge National Laboratory for several interesting and informative comments and suggestions during a short chat in May 1975. Our thanks also to Dr. F. G. Toman, Advanced Research Director, Nuclear Regulatory Commission for some comparative information on studies previously carried out in Panama. We thank Dr. William Bhajan, Terrestrial Ecology Division, RNC for acquainting us with several very important references cited herein and we acknowledge the

constructive commentary on the effects of background subtraction on Ge-ld spectra which we received from Professor Fausto Muñoz Ribadeneira of the University of Ecuador, Quito, Ecuador. Finally, we acknowledge the constructive guidance and criticism given to the preparation of this manuscript by Ms. Ana J. Correa and the detailed editing of Ms. Fern Galantai. ---Page Break--- Chapter 1 A, Introduction The radioisotope content of soil and vegetation samples in Puerto Rico on clearly defined transects was first measured by Kline (1968) and Kline et al. (1973). The accent of these studies was on the determination of radionuclide residuals which had fallen out or rained out on the land after the explosion of fission devices in the atmosphere. Among the hypotheses examined was the relationship between rainfall and radiomelic content (Odun et al. 1970) and a test of a hypothesis similar to the proposed "continental effect" (Roser and Cullen, 1965; Meyer et al., 1968). The following radionuclides, with the half-lives (T1/2) listed, were found in most samples: Ce-241/Pr, T1/2: 264 days/T2/2: 17.3 minutes; Cs-137/Ba-137, T1/2: 30.2 years/T1/2: 2.55 min.; and, rarely, Tl-208, T1/2: 65 days; T1/2: 35 days (Definition of chemical symbols given in Appendix B) In November, 1973, the Terrestrial Geology Program of the Puerto Rico Nuclear Center began a series of surveys in Barrio Telote, Arecibo, Puerto Rico. The objectives of these surveys were the development of sections of an environmental report of that area for the Puerto Rico Water Resources Authority. The entire report was used to comply with federal regulations governing the construction and operation of thermonuclear power generation facilities in the area. Some of the surveys were connected with the collection of background, radiological data such as the normal background levels of gamma activity (Block et al. 1975) and the identification of radionuclides already present in the proposed.

exclusion zone. Interestingly, Kline (1968) and Kline 2. (1973) do not appear to have studied soil radioactivity attributable to primordial radionuclides, even though it is estimated that 97% of natural

environmental background radioactivity to which man is continuously exposed is attributable to extinction by primordial terrestrial radionuclides (Kisty, 1974) and cosmic rays. Some radionuclides produced during atmospheric testing of fission or fusion devices can, however, easily enter the food chain leading to incorporation in the human body and their importance cannot be minimized. The cessation of atmospheric testing of strategic nuclear devices by the most technologically advanced nations (i.e. U.S.A. and U.S.S.R.) began with agreements in 1958 and continued by virtue of a treaty negotiated in 1963. This has led to a decline in environmental levels of some radionuclides, with T 1/2 less than years; some of which had been produced on a large scale since 1945. Nations which have developed nuclear arms capabilities more recently, such as France, People's Republic of China, and India, have concentrated, for the most part, on small but relatively high-yield devices. Most of these devices are generally believed not to have been of the megaton class and have probably not produced copious quantities of Pu-239. The purpose of this report is to present and interpret gamma-ray energy spectra measured on soil samples collected in the projected thermonuclear power plant exclusion zone in Barrio Islote. The interpretation consists of 2 chapters, in the first of which an identification of peak gamma energies with specific radionuclides will be made. The second chapter is devoted to estimating the soil burden of Cs-137 in Barrio Telote as a complement to estimates made by Kline (1968) and Kline et al. (1973). In addition, a Caveat Lector has been added for the purposes of cautioning the user of the data contained in Appendix A, concerning their inapplicability to petrology of the zone. B. Qualitative Survey 1.

Experimental Design

Some 19 samples of soil were collected at points on 3 transects within the area designated as the exclusion zone by the FRWRA. The area is shown in Map T and relates the position of the exclusion zone to coastal features of Puerto Rico and to other towns on the island. The zone is marked as "PROPOSED NORCO SITE". The transects were marked out from north to south through the eastern, central, and western sections of the zone. The zone is nominally bounded by the 66°37'22" and 66°36'30" west longitudes and the transects were nominally bounded by the 18°29'36" and 18°29'6" north latitudes. The proposed plant site has coordinates of approximately 66°36'57" west by 18°29'6" north. All samples were collected within essentially 1 km of the proposed plant site. The samples collected were all taken from the surface 6 inches. The soils of the site are the products of weathering of calcareous sandstones with the exception of the sugar cane field soils, most of which are organic soils (Bonnet and Roberts, 1967). Owing to the close proximity of the zone to the coast, these soils are probably salty and would be expected to contain radionuclides from the decay of Ra-226. Gamma spectra of the samples were taken using a lithium drifted germanium detector (GeLi) with a nominal efficiency of 1% operated at liquid nitrogen temperature. Typical counting time was 2000 minutes and a 4096 channel analyzer was used to store the pulse height information. The energy increment of the multi-channel analyzer was 1/2 KeV per channel using as an energy calibration peak the second peak of the Co-60 doublet at 1,332.51 KeV (Dyer and Bate, 1972). All soil samples were air dried and mechanically crushed before mounting over the detector in a Marinelli beaker of polystyrene construction.

2. Results and Discussion

In general, actual data on counts versus channel number, the approximate location at which samples were collected, and a description of the properties of each sample are given in

Appendix A. All samples exhibited nearly the same pattern of peaks and the richest spectra were

observed for samples designated ABBHDO3, ABOO16, and ABSCF2, which were clay, sandy loam, and sandy muck (Tiburones Muck; O.F. Bonnet and Roberts, 1967) respectively. Since the spectrum of ABBIDO3 was not only the most intense, but also the richest of all of the spectra, a plot was made of the log of the number of counts in each channel versus the energy of the channel to show clearly how the well-resolved spectrum appears. This plot is shown in Figs. I-VI. ---Page Break--- Primordial Radionuclides Peak assignments are based on the tacit assumption that the majority of the radionuclides encountered are primordial in origin or are descendants of primordial radionuclides (Kanasevich, 1968). The uranium (U-238) and thorium (Th-232) decay chains (decay series) shown in Figs. VII and VIII, respectively, are principally responsible for natural radioactivity, although K-40 is also a very important source of natural environmental gamma ray emissions. The energy of the gamma ray emitted by K-40 is relatively high (1460 keV); it is a relatively abundant radionuclide (0.01196 of all natural potassium is K-40); and about 12.36% of all K-40 disintegrations results in the emission of a gamma ray (Leighton, 1959). The fact that potassium is a micronutrient, essential for growth and reproduction of all animals and plants, establishes K-40 as a prime candidate for gamma ray induced mutagenicity. Small inputs to natural environmental radioactivity also come from the neptunium (Np-237) series (Fig. IX) and the actinium (Ac-227) series (Fig. X). The parent nuclides of the thorium, uranium, neptunium, and actinium series are Th-232, U-238, Np-237, and U-235, respectively, and the half-lives for each parent are 1.39 x 10^10, 4.51 x 10^9, 2.05 x 10^6, and 7.07 x 10^8 years respectively. The average isotopic ratio of U-236 to U-235 established for many different samples taken from the natural environment is about 138.5:1 (Leighton, 1958).

suggesting that gamma radiation from U-238 decay products is probably much more readily observable than gamma radiation from U-235 daughters (Bete, 1975). ---Page Break--- ---Page Break--- ---Page Break--- ney AB s0U3 os ox Lo on " ey siunoo 5) In 913 ---Page Break--- ---Page Break--- ---Page Break--- ---Page Break--- FIG VII (4n+2) ---Page Break--- FIG VIII | 4 ! Li og sw a ae a ---Page Break--- FIG IX ---Page Break--- FIG X Ted] ved BE st ss ey gs 33 | T et ey or er ar oF (ane3) ---Page Break--- Crustal abundance of each of the primordial parent compounds has been estimated by Taylor (1964), and shown, as might be expected from its much shorter half-life, that there is a negligible quantity of Pb-237 in the crust. Average crustal concentrations of 9.6, 2.7, and 0.019 ppm for U-232, U-238, and U-235 respectively were reported (Taylor, 1964). This suggests that Th-252 and its more recent daughters are probably easier to observe than U-238, though the older daughters of U-238 that are gamma emitters might be observed using the Ge-Li if long counting times are used. The probability of observing a gamma ray from U-235 or its progeny is probably low unless chemical or physical pre-concentration of uranium in the sample is carried out. The energy dependence of the efficiency of the Ge-Li detector is a particularly frustrating weakness when assays of gamma-emitting radiometrics are attempted. The drop-off in photo peak detection efficiency as the energy of the peak increases from 50 to 2000 KeV is rather variable and, in general, is non-linear nominally between 50 and 2000 KeV (Gordon et al., 1968). Indeed, for two Ge-Li detectors studied, photo peak efficiencies for 2000 KeV peaks were approximately 1/30 and 1/250 their respective values for the 50 KeV region (Gordon et al., 1968). The data also suggest that at higher energies (1 + 500 = 2000 KeV), rather long counting times and background subtraction or count rejection techniques ought to be applied. The first of these alternatives was used during some

quantitative work and counting times of 2000 minutes were the longest practical counting periods. These counting times are long enough to develop sufficiently objective peaks for daughters of primordial radio-nuclides which emit gamma rays with energies in excess of 500 KeV, and which

are expected to be present in the samples. Referring to Figs. I-VI, the energy peaks for Ra-226 correspond quite closely to literature values (Dyer and Bate, 1972; Delepov and Peker, 1961); in fact, the energies are always within ±1 KeV of the literature values. The following primordial radionuclide daughters are represented by energy peaks in the gamma spectra and are also attributed to the (I) series: Th-234 (T1/2 = 245 days), Po-210 (T1/2 = 26.8 minutes), Bi-214 (T1/2 = 29.7 minutes), and Pb-210 (T1/2 = 1.32 minutes). The inferred absence of series members Pa-234m (T1/2 = 6.75 hours) and U-234 (T1/2 = 2.3380 years) and Th-230 (T1/2 = 8.3 x 10^4 years) is explained as follows. Pa-234 and Pa-234m are both the same radionuclide, the lower case "m" indicating an excited nuclear energy state associated with the radionuclide. The (n+2) decay series shown in Fig. VET indicates that U-238 is formed upon neutron capture of U-235. The Pa-234 subsequently decays via beta decay to Pa-234m, the excited radionuclide. The Pa-234m can then either decay to the ground state, creating Pa-234 through an internal transition (IT), or it may decay directly to U-234 via beta emission. The direct decay to U-234 is most frequently observed; 99.87% of all of its disintegrations result in U-234. However, the efficiency of emission associated with gamma energies above 43.5 KeV is so small as to make these resonances unobservable in the experimental arrangements used. The Pa-234m formed from 0.136 of the Pa-234m via one beta decay can also produce U-234 via a beta emission, but the quantity of Pa-234 is so small that possible visible resonances are obscured by background and chemical separation.

this Isotope would be necessary for the spectrum to be visible using the experimental configuration employed in this work. 'A gamma energy resonance at 53.3 KeV attributable to U-234 probably is obscured due to metallic shielding used over the Ge-Li detector, limiting its detection to all but the most intense peaks near this energy. Another peak which could be attributed to U-234 at an energy of 120.9 KeV is not considered to be statistically reliable and was (subjectively) eliminated. Element ™-230 lacks intense gamma peak emission at any energy. Rationales similar to these apply to the apparent absence of gamma emission peaks due to Rn-222, Po-218, and Po-211. The ground state of B-210 does not emit observable gamma rays and Pb-210 does not exhibit strong gamma resonances in the region between 50 and 2000 KeV. As indicated in the decay scheme of Fig. VII, Pb-206 is formed from such small quantities with such a short half-life that it is unlikely that any peaks attributable to it would be distinguishable from background. ---Page Break--- Element At-218 is not an emitter of gamma rays and is formed in only small quantities which decay rather rapidly. There are gamma resonances which correspond to Tl-210 and the gamma peak at 795 KeV is quite intense (Lederer et al., 1967). Nevertheless, Tl-210 is formed by only 0.01% of the disintegrations of Bi-214, and positive identification does not seem warranted on this basis. Moreover, Tl-220 with a relatively short half-life does not build up and the peak assignment made for Tl-210 must be considered tentative. The daughters of Th-232 (the decay series) are clearly present, although an overlap of energies with members of the (decay) series obscure some quantitative aspects of the analysis. The following Th-232 positive identifications were made: Th-232, Ac-228, Ra-226, Tl-222, Bi-212, and Pb-208. Element Ra-228 has no intense gamma peak in the range of energies considered. There is a very slight indication of a peak at 542 KeV, indicative of Rn-220, but the subjectively drawn figure

does not show this clearly, and Rn-220 being a gas with T 1/2 = 55 seconds may not show up sharply prior to the central concentration of its precursor Ra-222. Also, only 0.01% of the disintegrations of Rn-200 result in gamma ray emission. The two radionuclides Po-216 and At-216 do not emit gamma rays upon disintegration. The radionuclide Po-212 (T 1/2 ≈ 36 seconds) is formed by a small percentage of beta emissions from Bi-222, the majority of the Bi-212 emissions

leading to Po-212 (T 1/2 ≈ 0.5 + 0.3 microseconds). Thus while Po-212 does emit a gamma ray of energy near 570 keV, it is not observed. ---Page Break--- c. Radionuclides Created by Fission and Fusion Devices Aside from some transuranium elements (i.e. Pu-239, Am-241, etc.), fission and some fusion devices produce few radionuclides having half-lives comparable to a generation of mankind. Of these few, a small percentage are considered particularly important because of their tendency toward accumulation in biota and "biological amplification." Among these elements Cs-137 (T 1/2 = 30 years), Ce-144 (T 1/2 = 284.4 days), Sb-125 (T 1/2 = 2.73 years), Sr-90 (T 1/2 = 28.9 years/64 hours) and others of comparable half-life are considered sufficiently important that complete monitoring capability for them is a requisite for construction and operation of thermonuclear power generation facilities. Constant ratios of K to Cs in biota (Fisenbud, 1963), the tendency for mammals to concentrate Sr-90 in bone and cartilage tissue (Rand Corp., 1953) and the deleterious effects of I-131 binding to the thyroid gland are thus recognized in legal requirements for pre-operational monitoring programs for nuclear power generation facilities (Puerto Rico Water Resources Authority, 1976). Wine (1968) and Kline et al. (1973) reported on radionuclides measured at a time when the USA-USSR atmospheric test-ban treaty had been in effect for about 15 years. They found the fission-produced radionuclides most easily determined using NaI (Tl) detection as mentioned in the Introduction.

The inspection of the gamma spectra in Appendix A by us revealed that Cs-137, which is produced in high yield by both fast and slow neutron source devices, both by U-235 and Pu-239. ---Page Break--- Tension, is noticeably present in all of the soil samples taken (Energy = 661.6 KeV). Radioisotopes which also are suspect are Sb-125 and Ce/Pr- neither of which can be established as present on a statistically reliable basis by the peaks corresponding to energies of gamma rays emitted by these. This identification for these two fission-fusion produced radionuclides is considered tentative. In the case of Sb-225, with T 1/2 = 2.7 years, many gamma rays are emitted upon disintegration, of which the most intense occur at energies of 427 and 598 KeV respectively. The case for Ce/Pr is weaker both because of its shorter half-life and the lack of prominence of either of the peaks ascribed to it (133.5 KeV and 80 KeV). The data do suggest that there should be a prominent Pr-141 peak at 697 KeV, but the peak lacks sufficient definition. The 80 KeV peak from the Ce radioisotope is considerably broadened by other, low energy peaks, one of which may be due to low energy resonance from heavier transuranic elements. Element, Pu-239 is tightly bound by soil, showing little propensity for transfer into plants or animals, and it is not easily introduced into the food chain (Bisenbud, 1963). There is no reason for suspecting its presence in ABEHDO3 or other samples however, since it is not easily detectable even with higher efficiency NaI (72) equipment (Burson, 197). The 129 KeV peak is interesting in that it is rather broad, implying that more than one radioisotope is contributing to its intensity. Furthermore, it is clearly seen from the background spectrum. Further work including chemical separation and re- counting should be used to establish what constituents are present in this peak and if any measurable level of Pu-239 is present since Pu-239 may be a future source of energy in nuclear power.

plants. ©. Effects on Biology The fact that the distribution of gamma energies in this area is nearly wholly dependent on primordial radionuclides coupled with the previously measured low background radioactivity (Block et al., 1975) would seem to rule out any unusual mutagenic activity attributable to radionuclides and localized in Barrio Islote. Fallout patterns would have no obvious climatological mechanics for concentration such as that postulated and observed by Osburn (1963) and Osawa et al. (1962). On the other hand, Osburn (1965) has pointed out that in cases where a high radioactivity exposure or radionuclide burden is coupled with other environmental stresses during radio-sensitive periods in the life cycle, some preferential uptake is possible and mutagenic

activity would be expected to increase. Also, the distribution or geographical range of a species of plant or animal may be affected by natural primordial radionuclide distributions. Anomalous morphological forms have been shown to be more prevalent in tundra species exposed to higher radiation dosages (Osburn, 1961; Osburn, 1963) and granite outcrops have been shown to have communities in which the distribution of plant species follows the radioactivity gradient in the outcrop (McCormick and Cotter, 196?). Insoluble humic acid or peat fractions in the upper layers of soils have been invoked as a geochemical enrichment factor for (UO2) (Szalay, 1964). Although such studies are complicated by the form and solubility of the cation under investigation, they ought to be enriched in these fractions at least as well as others. The plant census made in the Islote area has not revealed any unusual or marked trends with respect to changes in morphological form, although plant analysis at the cellular level has not been carried out. Periods of severe extended drought continuing for as long as 3-4 months might be considered important stress factors in the development of plant communities, particularly on the shallow sandy soils of

the Limestone formations. Under such conditions, a sudden increase in the natural background radiation level could produce observable changes in the morphology and population distribution. Genetic changes also ought to be accelerated, perhaps to observable levels within a few years: Chapter I Cestun - 137 Estimates One 1000 gm. soil sample, ABBPON for which the gamma spectrum using the Ge-Li detector had been taken, was subsequently doped with 100 pCi (nominal) of a commercial preparation of the radionuclide. The dopant Cs-137 was purchased from New England Nuclear, Boston Mass. 45 @ 1 mCi sample in approximately 0.5 M hydrochloric acid solution, with a listed radiochemical purity of greater than 99%. The contaminated soil (ABBIOM Cs) was then counted for the same period of time as the uncontaminated sample and a background subtraction was made. For this measurement, a subjective peak integration was employed. The interpretation was based upon the cesium peak characteristics demonstrated in Fig. TV, Chapter I. In general, the count maxima lay between two channels and in all spectra at an energy between 611 and 613 KeV. The integrated peak count included the two central channels and six channels on either side of these two. Since Cs-137 is present in the soil at a relatively low level, and the soil matrices were similar, the Cs-137 spike increment was considered sufficiently low that a linear extrapolation of the increment would give a reasonable estimate of the original Cs-137 burden. Estimates for other samples were based on the counts per Cs-137 increment in this sample as well, even though there may have been slight variations in the geometry of the counting configuration. The sample with the Cs-137 spike - ABBOL + - showed an increase of 27.3 counts per pCi of Cs-137 over the unspiked samples. The error in this incremental activity is estimated to be no worse than about 10% and this value was applied to each sample to calculate an activity per gram of sample. The calculated

Activities are shown in Table TI-1. Two of the samples: ARMTO2+ and ABOOLI+ were spiked with HCl to give enhanced K-K0 peaks compared with their unspiked counterparts ABMTOW and ABOOI4 respectively. The KCl used was of analytical purity and is considered free of Cs-137 contamination. Typical reproducibility of results from long Ge-Li counting can be inferred from the fact that the samples not spiked with KCl gave Cs-137 activities of 0.226 and 0.121 pCi/gm respectively, while the spiked samples exhibited Cs-137 activities of 0.125 and 0.186 pCi/gm suggesting that a more readily standardized method than this one ought to be developed unless an assay reproducibility of 35% can be tolerated in environmental monitoring of soil near a nuclear plant. An average Cs-137 activity with a calculated standard deviation (to show an approximate range of variability) was calculated excluding samples ABOOOY and ABBOOWL of which such a small quantity respectively was counted, that it was felt that a counting geometry equivalent to the

other samples was probably not conserved. This average Cs-137 activity was 0.131 pCi/gm with a distribution in values characterized by a standard deviation of 0.019 pCi/gm. Density measurements on the undried soil samples were not taken and direct comparison with the data of Kine et al. (1973) is not possible since that data was reported in terms of activity per unit surface area of soil.

Table TT Sample AST Activity in pCi/gm Assoowi* 0.138 ammo x0001 'aneow 030010 omscre 'ABTS 0.026 animes 'm0014 amO12 '83000 8 ABasL 'apooor* 'ABDOF2 -xBBCOL 'RBOOLI, +8003, AROS 'Values for these samples are given lower significance because the weight of sample used indicates that counting geometry may have been significantly different from other samples. AMMMOe+ is AMMTON with a known KCl spike ARCOM is ABOOLI with a known KCl spike.

Lector: On the use of Appendix A for inferences of the geological and petrological.

Development in the northern coastal zone of Puerto Rico. An area along the north coast extending eastward toward Dorado has been suggested as a possible petroleum exploration region. Puerto Rico is situated on the northeastern edge of the Caribbean plate, a reasonably stable seismic situation with a major earthquake repetition rate which is unknown. The last major earthquake associated with the submersion of the outlying Atlantic sea floor took place before recorded history and despite some minor tremors felt occasionally on the western and northern part of the island, Puerto Rico must be considered a less than likely place for the occurrence of a major tremor. Nevertheless, since the Caribbean plate is fairly well delineated and diving of the Atlantic sea floor has occurred on its eastern and northern (the Puerto Rico Trench) edge, there exists a stratigraphy in which petrologists are generally skeptical about the probability of encountering major petroleum deposits (Atwater, 1976). However, the physiography of the northeast site has been reviewed (Puerto Rico Water Resources Authority, 1974 b), so an important consideration for petroleum exploration. The Clenaga Tiburones, bounded by ridges of the Arecibo and Manatí rivers, is filled with approximately 30 meters of peat and organic-rich clay and is probably underlain by limestone deposits (Blaz, 1973). At least four Quaternary eolianite ridge lines occur on the north central coast, and it is believed that the entire eolianite sequence was formed during Pleistocene sea level lowering. Additionally, shallow reflection seismic data off the north coast near Arecibo indicate the presence of at least two alluvial fills, to about 60 meters below sea level. The following conclusions by Hassan (1973) and Hassan and Fossin (1975) suggest that limestone corings both from land and sea side should be examined with respect to the Th/E ratio as determined using gamma spectroscopy, when evaluating oil-bearing potential of rock.

substrate. In sedimentary rocks, thorium has a detrital origin and remains stable during alteration and subsequent diagenesis. Second, the Th/K ratio and the paramagnetic electronic resonance associated with organic phases show parallel trends. As mentioned previously, the humic acid or peat-layer mobilization of ionic species such as Th ought to be at least as good as Wo," barring widely different solubility. Certainly, Th is quite soluble in any aqueous phase and might be expected to be well-fixed in organic layers and retained during subsequent diagenesis. Hassan (1973) noted that Th is almost entirely concentrated in clay fractions and that it is believed to remain stable during alteration and diagenesis. This would mean that although enrichment in organic phases is entirely likely via exchange with humic acid, Th undergoes transfer to clay material during sedimentation and alteration. Hassan (1973) concurs with Seeley (1964) that uranium may be much more stable in organic and phosphate constituents. The Th/K ratio in Silurian Eocene clays associated with petroleum-bearing substrate was well correlated with Tilite

crystal unity, which in turn is associated with deep burial diagenesis (Hassan and Hossin, 1975). Thus, it is tempting to speculate upon the Th/K ratio in the Islot or other offshore areas in Puerto Rico. Such speculation based upon Appendix A of this report is very ill-advised. The samples for which gamma spectra were measured were entirely composed of surface soils. The potassium in these soils would have been subject to intense leaching due to their location in a subtropical zone. A high degree of leaching of K compared with Th would be expected, and the net result could be artificially high values of the Th/K ratio when compared with deep-buried sedimentary deposits.

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---Page Break--- APPENDIX A 'Appendix K consists partially of numerical print-out of records stored on the PDP-10 computer at the Mayaguez A. & M. University computer center: 110 blocks in 24 files, Disk B (DSKB), with designation: /250351, 2503167 ---Page Break--- 'Appendix A shows the actual numerical values of counts per channel for the different spectra taken. The channel numbers in the column on the extreme left have the number of counts specified by the first numerical value to the right of the channel number. Subsequent channel numbers and the number of counts in each are read horizontally from left to right. For example, in spectrum ABB:003 (Fig. A-1), channel number 104 registered 801 counts, channel number 105 registered 772 counts, channel number 106 registered 730 counts, etc. Background measurements were made frequently and counting times were of the order of 2000 minutes to develop major gamma peaks. These long counting times reflect the relatively low efficiency of Ge-14 as a detector of gamma emissions. Table AA-I gives basic data on each spectrum taken and shows on which page the spectrum is located. ---Page Break--- Table AA-D 'Time (min.) Level —— counting Designation watt simulation channel ---Page Break--- 4095 channels: ---Page Break--- ---Page Break--- a ABBHO03 count ---Page Break--- ---Page Break--- ABBHO03 count ---Page Break--- er measurements ---Page Break--- ABBHO03 count ---Page Break--- as ABBKO03 4095 channels ---Page Break--- ---Page Break--- 6 ABBKO03 count ---Page Break--- ---Page Break--- " ABBKOO3 count. a a 1 ' 4 1 a 3 ---Page Break--- aaa ---Page Break--- ABBKOO3 com ---Page Break--- Ld ABBOOW1 w.transect OWS CHANNELS 2838 EEEtE Etat ---Page Break--- ---Page Break--- re) ABBOOW1 count ---Page Break--- ---Page Break--- ABBOOW1 count 3 ---Page Break--- ---Page Break--- ABBOOW1 count CHANNEL NO. ---Page Break--- ry ABMT02 — 4095 channels ---Page Break--- ---Page Break--- 1

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